Classical theory of cylindrical nonlinear optics: Sum- and difference-frequency generation

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Classical theory of cylindrical nonlinear optics: Sum and difference frequency generation

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The process of two input electromagnetic waves at different frequencies overlap in a nonlinear medium is a typical phenomenon of nonlinear optics. The traditional method of dealing with such problem is utilizing coupled wave equations. In this paper, sum and difference frequency generation of cylindrical electromagnetic waves in a nonlinear medium has been investigated in a new way. We use exact solutions of Maxwell equations to describe the propagation of cylindrical electromagnetic waves in a nonlinear medium and show that sum and difference frequency generation comes out quite naturally from such exact solutions. For comparison, the traditional method of utilizing coupled wave equations is also discussed, and we find that the results obtained from two different approaches are consistent with each other.

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I. INTRODUCTION

Sum frequency generation (SFG) and difference frequency generation (DFG) are second-order nonlinear optical processes in which two input electromagnetic waves with different frequencies overlap in a nonlinear medium and generate an output wave at the sum or difference frequency of those of the applied fields[1–4]. As typical phenomena in nonlinear optics, SFG and DFG in a nonlinear medium have been widely studied and used in various fields. For example, the generation of tunable optical radiation typically relies on nonlinear frequency conversion in nonlinear medium [5–7], and as a noninvasive and noncontact probe of the electronic and structural properties, SFG has been frequently used in chemistry [8, 9], biochemistry [10, 11] and biophysical [12, 13]. On the subject of SFG and DFG, plane waves are always considered in most works. Features of SFG and DFG with cylindrical or spherical waves, however, remain poorly studied. In this paper, we will investigate SFG and DFG of cylindrical electromagnetic waves in a nonlinear medium.

Our motivations are twofold. Firstly, the previous study [14] on cylindrical nonlinear optics shows that the exact solution obtained in reference [15] can be used to describe second-harmonic generation well, while we want to go one step further here by extending the method to deal with other phenomena like SFG and DFG. Similarly to second-harmonic generation, SFG and DFG also come from the secondary nonlinear polarization of a nonlinear medium. Secondly, previous studies [14–17] of the exact solution usually focus on single electromagnetic wave propagation in a nonlinear medium. However, for the case of two or more waves existing in the nonlinear medium simultaneously, there are few effective methods to deal with it. Superposition of all the descriptions of the waves separately propagation in the nonlinear medium not leads the answer. The nonlinearity of the medium will cause interactions between cylindrical electromagnetic waves, which are traditionally described by a set of coupled wave equations. So it is interesting to give a method to extend the exact solution to deal with problems of interactions between the cylindrical electromagnetic waves.

There are some highlights in our work. For example, we give an effective method to deal with problems of interactions between the cylindrical electromagnetic waves, and we give an explicit analytical expression which contains all the main nonlinear optical effects. To the best of our knowledge, such expression is the first explicit analytical expression which contains all the main nonlinear optical effects. The traditional method describing nonlinear optical effects is the coupled-wave equation approach which can be solved only numerically in the cylindrical geometry. On the other hand, the present work gives a verification of previous work [14], which put forward some approximations to deal with SHG. However, it needs to verify that are these approximations applicative in other cases? The present work shows that these approximations are applicative at least in the case of discussing SFG and DFG.

In this article, we will employ two different methods to investigate SFG and DFG of cylindrical electromagnetic waves in a nonlinear nondispersive medium. Firstly, following the method proposed in reference [15] for constructing exact axisymmetric solutions of Maxwell equations in a nonlinear nondispersive medium and the method proposed in reference [14] for describing second-harmonic generation by the exact solutions, we will demonstrate that this exact axisymmetric solution, which has been successfully used to discuss electromagnetic shock waves [15] and second-harmonic generation [14], can also be used to describe SFG and DFG. This will be discussed in detail in Sec. II. Secondly, to verify effectiveness of the new method, we will use coupled wave equations, which are derived in reference [14] from imitating plane nonlinear optics and describing the interaction between cylindrical electromagnetic waves and a nonlinear medium, to study SFG and DFG of cylindrical electromagnetic waves. This will be discussed in detail in Sec. III. At last, we end our paper with a short summary in Sec. IV.

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II. ANALYSIS OF SFG AND DFG BY USING EXACT SOLUTION

A. analysis with simple approximation

At the beginning, we shall introduce the physical model we will discuss in this work. Considering the medium possesses an axis of symmetry and taken as the z axis of a cylindrical coordinate system (r, ϕ, z), we use the axisymmetric model in which the fields are independent of ϕ and z, then the Maxwell equations can be written as follows [15]:

$$\frac{\partial H}{\partial r} + \frac{H}{r} = \varepsilon(E)\frac{\partial E}{\partial t}, \qquad \frac{\partial E}{\partial r} = \mu_0 \frac{\partial H}{\partial t}, \tag{1}$$

where $H \equiv H_\phi(r,t)$, $E \equiv E_z(r,t)$, $\varepsilon(E) = \mathrm{d}D/\mathrm{d}E = \epsilon_0\varepsilon_1 \exp(\alpha E)$, with ε_1 and α are certain constants. Thus $P = D_0 + \epsilon_0(\varepsilon_1 - 1)E + \epsilon_0\varepsilon_1\alpha E^2/2 + \cdots$, and $\chi^{(2)} = \varepsilon_1\alpha/2$. Exact solution of such system can be written as [15]:

$$\begin{aligned} E &= \mathcal{E}(\rho e^{\alpha E/2}, \tau + \frac{Z_0\alpha\rho H}{2\sqrt{\varepsilon_1}}), \\ H &= \frac{\sqrt{\varepsilon_1}e^{\alpha E/2}}{Z_0}\mathcal{H}(\rho e^{\alpha E/2}, \tau + \frac{Z_0\alpha\rho H}{2\sqrt{\varepsilon_1}}), \end{aligned} \tag{2}$$

where $\mathcal{E}(\rho,\tau)$ and $\mathcal{H}(\rho,\tau)$ represent the solution of linear problem (1) with $\alpha = 0$, $\rho = r/a$, $\tau = t/(\sqrt{\epsilon_0\varepsilon_1\mu_0}a)$, $Z_0 = \sqrt{\mu_0/\epsilon_0}$ and a is a constant with the dimension of length.

Now we will use Eq. (2) to derive exact solution of two cylindrical electromagnetic waves propagation in an infinite nonlinear medium. The solution of single cylindrical wave propagation in an infinite and linear medium is: $\mathcal{E}(r,t) = \zeta J_0(kr)\cos\varpi t$ and $\mathcal{H}(r,t) = -\zeta J_1(kr)\sin\varpi t$. Here J_m is a Bessel function of the first kind of order m, ζ is a constant and $k = \varpi\sqrt{\epsilon_0\varepsilon_1\mu_0}$. For linear medium, superposition principle is always applicable. The exact solution can be easily extended to describe two cylindrical waves with frequencies ϖ_1 and ϖ_2 propagation in the linear medium:

$$\begin{aligned} \mathcal{E}(r,t) &= \zeta_1 J_0(k_1 r)\cos\varpi_1 t + \zeta_2 J_0(k_2 r)\cos\varpi_2 t, \\ \mathcal{H}(r,t) &= -\zeta_1 J_1(k_1 r)\sin\varpi_1 t - \zeta_2 J_1(k_2 r)\sin\varpi_2 t. \end{aligned} \tag{3}$$

Rewriting it in variable (ρ, τ), the solution becomes:

$$\begin{aligned} \mathcal{E}(\rho,\tau) &= \zeta_1 J_0(k_1\rho a)\cos(\varpi_1\tau\sqrt{\epsilon_0\varepsilon_1\mu_0}a) \\ &\quad + \zeta_2 J_0(k_2\rho a)\cos(\varpi_2\tau\sqrt{\epsilon_0\varepsilon_1\mu_0}a), \\ \mathcal{H}(\rho,\tau) &= -\zeta_1 J_1(k_1\rho a)\sin(\varpi_1\tau\sqrt{\epsilon_0\varepsilon_1\mu_0}a) \\ &\quad - \zeta_2 J_1(k_2\rho a)\sin(\varpi_2\tau\sqrt{\epsilon_0\varepsilon_1\mu_0}a). \end{aligned} \tag{4}$$

By using Eq. (2) we can obtain the solution of nonlinear problem:

$$\begin{aligned} E &= \zeta_1 J_0(k_1 r e^{\alpha E/2})\cos(\varpi_1 t + \alpha\mu_0\varpi_1 rH/2) \\ &\quad + \zeta_2 J_0(k_2 r e^{\alpha E/2})\cos(\varpi_2 t + \alpha\mu_0\varpi_2 rH/2), \end{aligned} \tag{5}$$

$$\begin{aligned} H &= -\frac{\sqrt{\varepsilon_1}e^{\alpha E/2}}{Z_0}\Big(\zeta_1 J_1(k_1 r e^{\alpha E/2})\sin(\varpi_1 t + \alpha\mu_0\varpi_1 rH/2) \\ &\quad - \zeta_2 J_1(k_2 r e^{\alpha E/2})\sin(\varpi_2 t + \alpha\mu_0\varpi_2 rH/2)\Big). \end{aligned} \tag{6}$$

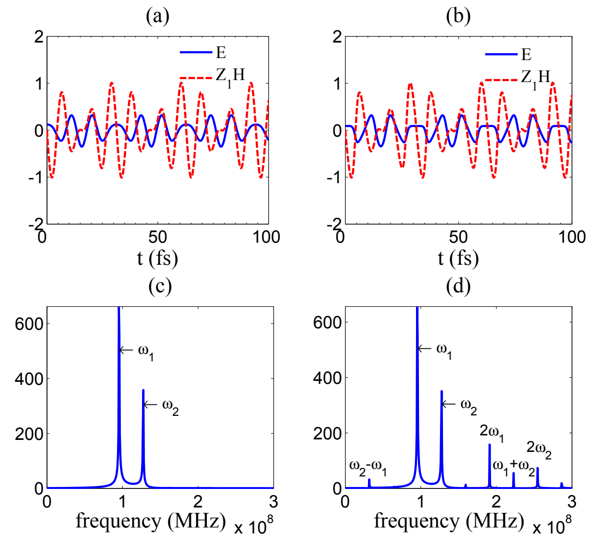

FIG. 1: (Color online) (a) Calculation result of oscillograms of two cylindrical electromagnetic waves propagation in an infinite linear medium by using exact solution (5,6) with $\alpha = 0$. We use $\zeta_1 = \zeta_2 = 1$, $r = 0.7\ \mu$m, $\varpi_1 = 6\times10^8$ MHz, $\varpi_2 = 8\times10^8$ MHz and $Z_1 = \sqrt{\mu_0/(\epsilon_0\varepsilon_1)}$. (b) Calculation result of oscillograms of two cylindrical electromagnetic waves propagation in an infinite nonlinear medium by using exact solution (5,6) with $\alpha = 0.3$. Other parameters remain the same as Fig. 1(a). (c) Frequency spectrum of the electric field when two cylindrical electromagnetic waves propagation in an infinite linear medium. The calculation result is based on Fig. 1(a). (d) Frequency spectrum of the electric field when two cylindrical electromagnetic waves propagation in an infinite linear medium. The calculation result is based on Fig. 1(b).

This solution describes two cylindrical electromagnetic waves propagation in a nonlinear medium and one can obtain the full information of E and H varies with r and t from it. For example, if we observe E at $r = 0$, we can get:

$$E = \zeta_1\cos(\varpi_1 t) + \zeta_2\cos(\varpi_2 t). \tag{7}$$

The exact solution (5,6) shows that the electric field and magnetic field of the cylindrical electromagnetic waves in a nonlinear medium are not separate, but coupling with each other by nonlinear coefficient α. Such coupling effect will change the frequency spectrums. Figure 1 shows the differences of frequency characteristics of two cylindrical electromagnetic waves propagation in an infinite linear and nonlinear medium. When two cylindrical electromagnetic waves with frequencies ϖ_1 and ϖ_2 propagate in the linear medium, the frequency spectrum of the electric field only have the two base frequencies ϖ_1 and ϖ_2, which is shown in Fig. 1(c). However, when two cylindrical electromagnetic waves propagate in the nonlinear medium, as shown in Fig. 1(d), the frequency spectrum of the electric field contains not only two base frequencies ϖ_1 and ϖ_2, but also other frequencies, such as $2\varpi_1$, $2\varpi_2$, $\varpi_1+\varpi_2$ and $\varpi_1-\varpi_2$. The calculations are based on the exact solution (5,6). It implies that the exact solution may be used to deal with problems of SFG and DFG. Reference [14] has proposed a method to derive second-harmonic generation from Eq. (2).

Here we want to go one step further by extending the method to deal with problems of SFG and DFG.

Similar to reference [14], we begin our discussion by using the approximation $\alpha\mu_0\varpi rH \ll 1$ and $\exp(\alpha E/2) \approx 1$, and writing the magnitude of the magnetic field as:

$$H \approx \gamma_1 \sin\varpi_1 t + \gamma_2 \sin\varpi_2 t, \tag{8}$$

where $\gamma_i = -\zeta_i\sqrt{\varepsilon_1}J_1(k_i r)/Z_0, i = 1,2$. Substitution Eq. (8) into Eq. (5) leads

$$\begin{aligned} E &\approx \zeta_1 J_0(k_1 r)\cos(\varpi_1 t + x_{11}\sin\varpi_1 t + x_{12}\sin\varpi_2 t) \\ &\quad +\zeta_2 J_0(k_2 r)\cos(\varpi_2 t + x_{21}\sin\varpi_1 t + x_{22}\sin\varpi_2 t), \end{aligned} \tag{9}$$

where $x_{ij} = \alpha\mu_0 r\varpi_i\gamma_j/2 = -\alpha r k_i\zeta_j J_1(k_j r)/2, i, j = 1,2$. Using $\alpha\mu_0\varpi rH \ll 1$, viz. $x_{ij} \ll 1$, $\cos(x_{ij}\sin\varpi_i t) \approx 1$, and $\sin(x_{ij}\sin\varpi_i t) \approx x_{ij}\sin\varpi_i t$, we can simplify Eq. (9) as follows:

$$\begin{aligned} E \approx\ & \underbrace{-\frac{x_{11}\zeta_1 J_0(k_1 r) + x_{22}\zeta_2 J_0(k_2 r)}{2}}_{\text{optical rectification}} + \underbrace{\zeta_1 J_0(k_1 r)\cos\varpi_1 t}_{\text{base frequency of } \varpi_1} + \underbrace{\zeta_2 J_0(k_2 r)\cos\varpi_2 t}_{\text{base frequency of } \varpi_2} + \underbrace{\frac{x_{11}\zeta_1 J_0(k_1 r)}{2}\cos 2\varpi_1 t}_{\text{second-harmonic of } \varpi_1} \\ & + \underbrace{\frac{x_{22}\zeta_2 J_0(k_2 r)}{2}\cos 2\varpi_2 t}_{\text{second-harmonic of } \varpi_2} + \frac{x_{12}\zeta_1 J_0(k_1 r) + x_{21}\zeta_2 J_0(k_2 r)}{2}\Big(\underbrace{\cos(\varpi_1 + \varpi_2)t}_{\text{sum frequency}} - \underbrace{\cos(\varpi_1 - \varpi_2)t}_{\text{difference frequency}}\Big). \end{aligned} \tag{10}$$

It implies that if two waves with frequencies ϖ_1 and ϖ_2 propagate in the nonlinear medium, there are respective second-harmonic, sum frequency and difference frequency generation. Second-harmonics of ϖ_1 and ϖ_2 have been studied in reference [14] and in this article we are interested in the term of SFG and DFG.

Equation (10) shows that sum and difference frequencies have the same amplitude but inverse direction of vibration. The amplitude of sum or difference frequency A_{sum} can be easily obtained as:

$$\begin{aligned} A_{\text{sum}} &= \frac{x_{12}\zeta_1 J_0(k_1 r) + x_{21}\zeta_2 J_0(k_2 r)}{2} \\ &= \frac{\alpha\zeta_1\zeta_2}{4}\Big(k_1 r J_0(k_1 r)J_1(k_2 r) + k_2 r J_0(k_2 r)J_1(k_1 r)\Big). \end{aligned} \tag{11}$$

Defining η_1 as the ratio between amplitudes of the sum frequency $\varpi_1 + \varpi_2$ and the base frequency ϖ_1 and η_2 as the ratio between amplitudes of the difference frequency $\varpi_1 - \varpi_2$ and the base frequency ϖ_1 in frequency spectrogram, we then have $\eta_1 = \eta_2$ and

$$\eta_1 = \frac{\alpha\zeta_2}{4}\Big(k_1 r J_1(k_2 r) + k_2 r J_0(k_2 r)J_1(k_1 r)/J_0(k_1 r)\Big). \tag{12}$$

Considering $\zeta_1 = 0$ or $\zeta_2 = 0$, viz. only one wave propagating in the nonlinear medium, equation (11) indicates that there is no sum and difference frequency. Simultaneously, equation (10) degenerates into the case of second-harmonic generation, which is the same as prevenient results [14].

B. analysis with improved approximation

As an approximate solution, equation (10) has shown most of the nonlinear optical phenomena, however, still need to be improved. There are several reasons. First, the amplitudes of SFG and DFG obtained by Eq. (10) are not in good agreement with exact solution (5,6). Figure 2(a) shows the efficiencies of generation of sum frequency (η_1) and difference frequency (η_2) with different r which ranges from 0 to 2. There are many differences between curves of using exact solution (5,6) and curves of using approximate solution Eq. (10). Second, equation (10) shows that the sum and difference frequency have the same amplitude, which turn out to be imprecise. Figure 2(a) shows that there are many differences between the amplitudes of the sum and difference frequency. Thus, the approximate solution Eq. (10) is not a very good approximation to describe SFG and DFG and some of the approximations, which are used to deduce the approximate solution Eq. (10), need to be improved.

Using numerical simulation we verify that Eq. (8) is a good approximation and the errors of Eq. (10) mainly arise from the approximation $\exp(\alpha E/2) \approx 1$, precisely, $J_0(kre^{\alpha E/2}) \approx J_0(kr)$. Such case also exists in the study of second-harmonic generation. Reference [14] introduces a correction factor to describe the feature of second-harmonic generation more precisely and gives an advanced approximation to show the origin of the correction factor. In what follows we will use the improved approximation proposed in reference [14] to replace the approximation $J_0(kre^{\alpha E/2}) \approx J_0(kr)$ and show that the improved approximation can also be used to deal with the problem of SFG and DFG.

Now we turn to deal with the problem of sum and difference

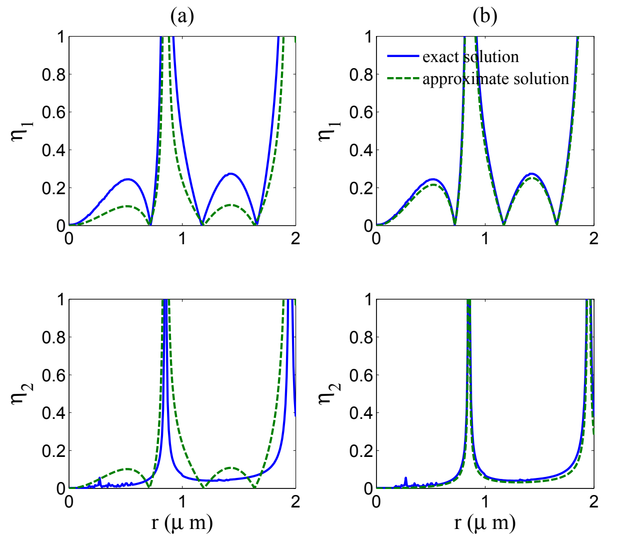

FIG. 2: (Color online) Comparison diagram of using exact solution and two approximate solutions. We use $\alpha = 0.3$, $\zeta_1 = \zeta_2 = 1$, $\varpi_1 = 6\times 10^8$ MHz and $\varpi_2 = 8\times 10^8$ MHz. (a) Calculation result of η_1 and η_2 by using exact solution (5,6) and approximate solution Eq. (10). (b) Calculation result of η_1 and η_2 by using exact solution (5,6) and approximate solution Eq. (17).

frequency by using Eq. (8) and the new approximation

$$J_0(kre^{\alpha E/2}) \approx J_0(kr)\Big(1-\frac{\alpha EkrJ_1(kr)}{2J_0(kr)}\Big). \qquad (13)$$

Substitution Eq. (13) into Eq. (5) leads:

$$\begin{aligned} E &= \zeta_1 J_0(k_1 r)\Big(1-\frac{\alpha Ek_1 rJ_1(k_1 r)}{2J_0(k_1 r)}\Big) \\ &\quad\times\cos(\varpi_1 t + x_{11}\sin\varpi_1 t + x_{12}\sin\varpi_2 t) \\ &\quad+\zeta_2 J_0(k_2 r)\Big(1-\frac{\alpha Ek_2 rJ_1(k_2 r)}{2J_0(k_2 r)}\Big) \\ &\quad\times\cos(\varpi_2 t + x_{21}\sin\varpi_1 t + x_{22}\sin\varpi_2 t). \qquad (14)\end{aligned}$$

We also can write Eq. (14) as the form of $E \approx E_0 + E_c$, where E_0 is the previous approximate solution Eq. (10) and E_c is a correction term. Here we focus on E_c, especially the correction factor of SFG and DFG. From Eq. (14) we can obtain E_c as:

$$\begin{aligned} E_c &= x_{11}E\cos(\varpi_1 t + x_{11}\sin\varpi_1 t + x_{12}\sin\varpi_2 t) \\ &\quad+x_{22}E\cos(\varpi_2 t + x_{21}\sin\varpi_1 t + x_{22}\sin\varpi_2 t). \qquad (15)\end{aligned}$$

Substitution $E = E_0$ into E_c and ignoring higher harmonics, we have:

$$\begin{aligned} E_c &= \frac{x_{11}A_1 + x_{22}A_2}{2} + x_{11}A_0\cos\varpi_1 t + x_{22}A_0\cos\varpi_2 t \\ &\quad+\frac{x_{11}A_1}{2}\cos 2\varpi_1 t + \frac{x_{22}A_2}{2}\cos 2\varpi_2 t + \frac{x_{11}A_2 + x_{22}A_1}{2} \\ &\quad\times\Big(\cos(\varpi_1+\varpi_2)t + \cos(\varpi_1-\varpi_2)t\Big), \qquad (16)\end{aligned}$$

where $A_0 = -(x_{11}\zeta_1 J_0(k_1 r) + x_{22}\zeta_2 J_0(k_2 r))/2$, $A_1 = \zeta_1 J_0(k_1 r)$ and $A_2 = \zeta_2 J_0(k_2 r)$. $E \approx E_0 + E_c$ leads a new approximate solution of the exact solution Eq. (5):

$$\begin{aligned} E \approx\ & \underbrace{(A_1 + x_{11}A_0)\cos\varpi_1 t}_{\text{base frequency of } \varpi_1} + \underbrace{(A_2 + x_{22}A_0)\cos\varpi_2 t}_{\text{base frequency of } \varpi_2} + \underbrace{x_{11}A_1\cos 2\varpi_1 t}_{\text{second-harmonic of } \varpi_1} + \underbrace{x_{22}A_2\cos 2\varpi_2 t}_{\text{second-harmonic of } \varpi_2} \\ & + \underbrace{\frac{x_{11}A_2 + x_{22}A_1 + x_{12}A_1 + x_{21}A_2}{2}\cos(\varpi_1+\varpi_2)t}_{\text{sum frequency}} + \underbrace{\frac{x_{11}A_2 + x_{22}A_1 - x_{12}A_1 - x_{21}A_2}{2}\cos(\varpi_1-\varpi_2)t}_{\text{difference frequency}}. \qquad (17)\end{aligned}$$

This approximate solution is better than Eq. (10) and two disadvantages, which are mentioned at the beginning of this section, have eliminated naturally here. Figure 2(b) shows the efficiencies of generation of sum frequency (η_1) and difference frequency (η_2) with different r which ranges from 0 to 2. We can find that curves of using exact solution (5,6) are in good agreement with curves of using approximate solution Eq. (17). Equation (17) shows that sum and difference frequency have different amplitudes, which can be observed from Fig. 2(b).

III. ANALYSIS OF SFG AND DFG BY USING COUPLED WAVE EQUATIONS

In what follows, we will use coupled wave equations of cylindrical electromagnetic waves interacting with a nonlinear medium, which is a traditional method, to deal with the problem of SFG and DFG from another perspective. The coupled wave equations have been deduced in reference [14] as follows:

$$\frac{\partial^2 E(\varpi_i)}{\partial r^2} + \frac{1}{r}\frac{\partial E(\varpi_i)}{\partial r} + k_i^2 E(\varpi_i) = -\mu_0\varpi_i^2 P_{NL}(\varpi_q = \varpi_i). \qquad (18)$$

P_{NL} is used as secondary nonlinear polarization $P^{(2)}$ for the problem of sum and difference frequency:

$$\begin{aligned}P_{NL}(\varpi_q=\varpi_1)&=2\epsilon_0\chi^{(2)}(-\varpi_1,\varpi_3,-\varpi_2):E(\varpi_3)E^*(\varpi_2)\\&=2\epsilon_0\chi^{(2)}(-\varpi_1,\varpi_2,\varpi_4):E(\varpi_2)E(\varpi_4),\\P_{NL}(\varpi_q=\varpi_2)&=2\epsilon_0\chi^{(2)}(-\varpi_2,\varpi_1,-\varpi_4):E(\varpi_1)E^*(\varpi_4)\\&=2\epsilon_0\chi^{(2)}(-\varpi_2,\varpi_3,-\varpi_1):E(\varpi_3)E^*(\varpi_1),\\P_{NL}(\varpi_q=\varpi_3)&=2\epsilon_0\chi^{(2)}(-\varpi_3,\varpi_1,\varpi_2):E(\varpi_1)E(\varpi_2),\\P_{NL}(\varpi_q=\varpi_4)&=2\epsilon_0\chi^{(2)}(-\varpi_4,\varpi_1,-\varpi_2):E(\varpi_1)E^*(\varpi_2),\end{aligned}\tag{19}$$

where $\varpi_3=\varpi_1+\varpi_2$, $\varpi_4=\varpi_1-\varpi_2$. Substitution of Eqs. (19) into Eq. (18) leads to two sets of coupling equations: one describes the coupling between $E(\varpi_1)$, $E(\varpi_2)$ and $E(\varpi_1+\varpi_2)$, hereafter we call it sum frequency coupling equations; the other describes the coupling between $E(\varpi_1)$, $E(\varpi_2)$ and $E(\varpi_1-\varpi_2)$ which is called difference frequency coupling equations. The sum frequency coupling equations can be obtained as:

$$\begin{aligned}\frac{\partial^2E(\varpi_1)}{\partial r^2}+\frac{1}{r}\frac{\partial E(\varpi_1)}{\partial r}+k_1^2E(\varpi_1)&=-2K_1E(\varpi_3)E^*(\varpi_2),\\\frac{\partial^2E(\varpi_2)}{\partial r^2}+\frac{1}{r}\frac{\partial E(\varpi_2)}{\partial r}+k_2^2E(\varpi_2)&=-2K_2E^*(\varpi_1)E(\varpi_3),\\\frac{\partial^2E(\varpi_3)}{\partial r^2}+\frac{1}{r}\frac{\partial E(\varpi_3)}{\partial r}+k_3^2E(\varpi_3)&=-2K_3E(\varpi_1)E(\varpi_2),\end{aligned}\tag{20}$$

where $K_j=\epsilon_0\mu_0\varpi_j^2d_{\text{eff}}$ with d_{eff} being effective nonlinear optical coefficient of the nonlinear medium. Below are difference frequency coupling equations:

$$\begin{aligned}\frac{\partial^2E(\varpi_1)}{\partial r^2}+\frac{1}{r}\frac{\partial E(\varpi_1)}{\partial r}+k_1^2E(\varpi_1)&=-2K_1E(\varpi_4)E(\varpi_2),\\\frac{\partial^2E(\varpi_2)}{\partial r^2}+\frac{1}{r}\frac{\partial E(\varpi_2)}{\partial r}+k_2^2E(\varpi_2)&=-2K_2E(\varpi_1)E^*(\varpi_4),\\\frac{\partial^2E(\varpi_4)}{\partial r^2}+\frac{1}{r}\frac{\partial E(\varpi_4)}{\partial r}+k_4^2E(\varpi_4)&=-2K_4E(\varpi_1)E^*(\varpi_2).\end{aligned}\tag{21}$$

Here $E(\varpi_j)$ is a function of r and t. Consider $E(\varpi_j)$ can be write as $E(\varpi_j)=A_j(r)J_0(k_jr)\exp(-i\varpi_jt)$, viz. the amplitude of the cylindrical electromagnetic waves only varying with r, then we can obtain:

$$\begin{aligned}\frac{d^2E_0(\varpi_1)}{dr^2}+\frac{1}{r}\frac{dE_0(\varpi_1)}{dr}+k_1^2E_0(\varpi_1)&=-2K_1E_0(\varpi_3)E_0^*(\varpi_2),\\\frac{d^2E_0(\varpi_2)}{dr^2}+\frac{1}{r}\frac{dE_0(\varpi_2)}{dr}+k_2^2E_0(\varpi_2)&=-2K_2E_0^*(\varpi_1)E_0(\varpi_3),\\\frac{d^2E_0(\varpi_3)}{dr^2}+\frac{1}{r}\frac{dE_0(\varpi_3)}{dr}+k_3^2E_0(\varpi_3)&=-2K_3E_0(\varpi_1)E_0(\varpi_2),\end{aligned}\tag{22}$$

and

$$\begin{aligned}\frac{d^2E_0(\varpi_1)}{dr^2}+\frac{1}{r}\frac{dE_0(\varpi_1)}{dr}+k_1^2E_0(\varpi_1)&=-2K_1E_0(\varpi_4)E_0(\varpi_2),\\\frac{d^2E_0(\varpi_2)}{dr^2}+\frac{1}{r}\frac{dE_0(\varpi_2)}{dr}+k_2^2E_0(\varpi_2)&=-2K_2E_0(\varpi_1)E_0^*(\varpi_4),\\\frac{d^2E_0(\varpi_4)}{dr^2}+\frac{1}{r}\frac{dE_0(\varpi_4)}{dr}+k_4^2E_0(\varpi_4)&=-2K_4E_0(\varpi_1)E_0^*(\varpi_2),\end{aligned}\tag{23}$$

where $E_0(\varpi_j)=A_j(r)J_0(k_jr)$. Equation (22) describes the process of sum frequency while Eq. (23) describes the process of difference frequency.

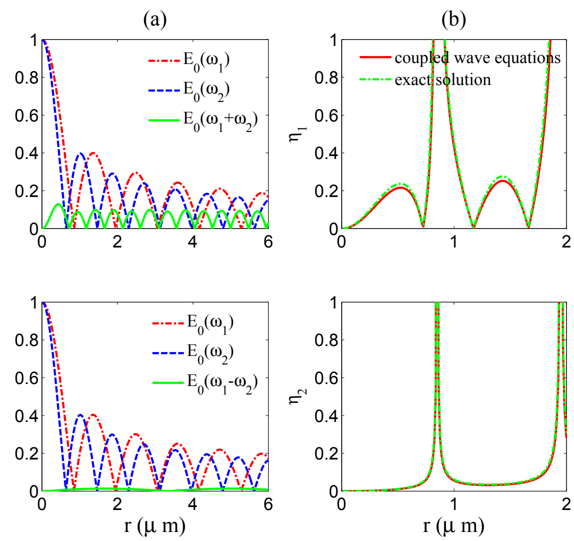

FIG. 3: (Color online) Calculation results of sum frequency and difference frequency generation by using coupled wave equations and exact solution. We use $d_{\text{eff}}=0.15$, $\alpha=0.3$, $\zeta_1=\zeta_2=1$, $\varpi_1=6\times10^8$ MHz and $\varpi_2=8\times10^8$ MHz. (a) The figure shows $E_0(\varpi_1)$, $E_0(\varpi_2)$ and $E_0(\varpi_1+\varpi_2)$ (above) and $E_0(\varpi_1)$, $E_0(\varpi_2)$ and $E_0(\varpi_1-\varpi_2)$ (below) as functions of r. The results are obtained by using coupled wave equations. (b) Efficiencies of generation of sum frequency (η_1) and difference frequency (η_2) with different r which ranges from 0 to 2 μm. The results are obtained by two methods: one is using coupled wave equations (solid curves) and the other is using exact solution (dashed curves).

Figure 3 shows calculation results of SFG and DFG by using coupled wave equations (22) and (23) and exact solution (5,6). The relation between d_{eff} and α is $d_{\text{eff}}=\alpha/2$, which has been obtained in previous work [14]. We use $d_{\text{eff}}=0.15$, $\alpha=0.3$, $\zeta_1=\zeta_2=1$, $\varpi_1=6\times10^8$ MHz and $\varpi_2=8\times10^8$ MHz. Figure 3(a) shows $E_0(\varpi_1)$, $E_0(\varpi_2)$ and $E_0(\varpi_1+\varpi_2)$ (above) and $E_0(\varpi_1)$, $E_0(\varpi_2)$ and $E_0(\varpi_1-\varpi_2)$ (below) as functions of r. It is obvious in Fig. 3(a) that the amplitude of sum frequency is very different from difference frequency. In the present case, the amplitude of sum frequency is much large than the amplitude of difference frequency. Figure 3(b) shows calculation results of efficiencies of generation of sum frequency (η_1) and difference frequency (η_2). From Fig. 3(b) we can find that descriptions of SFG and DFG by coupled wave equations are in good agreement with exact solution.

An important issue with SFG and DFG is that of phase-matching. In the present work, there is no discussion of phase-matching is because phase-matching of cylindrical nonlinear optics requires that the medium is inhomogeneous or dispersive. For more details one can see reference [14]. Here we focus on the propagation of cylindrical electromagnetic waves in a nonlinear and homogeneous medium, which the exact solutions have been obtained. And we find that the SFG and DFG come out quite naturally from such exact solution and the results obtained from exact solution are consistent with the results calculated by using coupled wave equations.

IV. CONCLUSION

In conclusion, we have used two methods to deal with the problem of cylindrical sum frequency and difference frequency generation. One method is using the exact solution obtained recently. We have found a simple method to deduce sum frequency and difference frequency generation from this exact solution. The other method is using traditional coupled wave equations. We have set up coupled wave equations of cylindrical electromagnetic waves interacting with nonlinear medium to describe sum frequency and difference frequency generation. Using the coupled wave equations we have analyzed features of cylindrical sum frequency and difference frequency generation, and found that the results are in good agreement with which are obtained by using the exact solution. Our results show that both methods are useful in dealing with the problem of cylindrical sum frequency and difference frequency generation and both have advantages in some aspects.

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[1] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).
[2] N. Bloembergen and P. S. Pershan, Phys. Rev. **128**, 606 (1962).
[3] N. Bloembergen, Rev. Mod. Phys. **54**, 685 (1982).
[4] Y. R. Shen, *Principles of Nonlinear Optics* (Wiley, New York, 1984).
[5] Y. Nakayama, P. J. Pauzauskie, A. Radenovic, R. M. Onorato, R. J. Saykally, J. Liphardt, and P. Yang, Nature **447**, 1098 (2007).
[6] M. A. Belkin, F. Capasso, A. Belyanin, D. L. Sivco, A. Y. Cho, D. C. Oakley, C. J. Vineis, and G. W. Turner, Nature Photonics **1**, 288 (2007).
[7] H. Suchowski, D. Oron, A. Arie, and Y. Silberberg, Phys. Rev. A **78**, 063821 (2008).
[8] L. Fu, G. Ma, and E. C. Y. Yan, J. Am. Chem. Soc. **132**, 5405 (2010).
[9] N. Ji, V. Ostroverkhov, C. S. Tian, and Y. R. Shen, Phys. Rev. Lett. **100**, 096102 (2008).
[10] Y. R. Shen, Nature **337**, 519 (1989).
[11] T. C. Anglin and J. C. Conboy, Biochemistry **48**, 10220 (2009).
[12] Yan Fu, H. Wang, R. Shi, and J.-X. Cheng, Biophysical Journal **92**, 3251 (2007).
[13] X. Lu, N. Shephard, J. Han, G. Xue, and Z. Chen, Macromolecules **41**, 8770 (2008).
[14] H. Xiong, L.-G. Si, J. F. Guo, X.-Y. Lü, and X. Yang, Phys. Rev. A **83**, 063845 (2011).
[15] E. Y. Petrov and A. V. Kudrin, Phys. Rev. Lett. **104**, 190404 (2010).
[16] H. Xiong, L.-G. Si, P. Huang, and X. Yang, Phys. Rev. E **82**, 057602 (2010).
[17] V. A. Es'kin, A. V. Kudrin, and E. Y. Petrov, Phys. Rev. E **83**, 067602 (2011).
[18] H.-S. Chan, Z.-M. Hsieh, W.-H. Liang, A. H. Kung, C.-K. Lee, C.-J. Lai, R.-P. Pan, and L.-H. Peng, Science **331**, 1165 (2011).